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May 11, 2006

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Abstract

The formation and evolution of stars and galaxies is a complex process that involves the cooling and collapse of dense interstellar clouds as well as energetic feedback on these clouds. Interstellar dust grains are central to the radiative transfer, thermal balance, and molecular processes in these clouds and can provide an important diagnostic. Hence, the effects of energetic processing of interstellar dust may have significant consequences. This may be studied in our own Galaxy, where observations have shown that an appreciable fraction of silicates formed in the outflows from red giants and supergiants have a crystalline structure. Yet, the fraction of crystalline silicates in the interstellar medium is very small, pointing towards an efficient crystalline-to-amorphous conversion process. Here we report experimental and modeling results that show that relatively

"low" energy (0.1 – 5.0 GeV) heavy ion cosmic rays can rapidly (~70 Million yrs) amorphize crystalline silicate grains ejected by stars into the interstellar medium. The implications of this are briefly discussed. We also examine the effects of cosmic ray processing of silicates in the solar system and in stellar debris disks. In the latter systems, cosmic ray processing may play a role for grains trapped in resonance with planetary companions. We speculate that energetic processing of interstellar dust is likely to be even more important in star forming galaxies, which have higher cosmic ray fluxes due to their much larger star formation rates and their emerging active black holes with associated jets.

1. Introduction

In forming galaxies, the interstellar medium (ISM) is exposed to UV and X-ray radiation from hot stars and active Super Massive Black Holes (SMBHs), cosmic rays (CRs) accelerated by supernovae, gamma-ray bursts and SMBH jets, and by shocks and ISM dust grain-grain collisions associated with these phenomena. This energetic feedback will significantly affect the star formation efficiency in such galaxies and may even control the growth of galaxies and their central super-massive black holes (Di Matteo, Springel & Hernquist 2005; Silk & Rees 1998). Most studies of feedback mechanisms in galaxy formation scenarios concentrate on the heating and outflow of ISM gas which truncate the star formation process (*negative* feedback). However, given the essential role that dust grains play during the star formation process (Draine 2003) it is equally important to examine the effects of energetic feedback on solid materials, especially of high energy particles because of their large penetration depths in star-forming molecular clouds.

Interstellar dust grains, while comprising only ~1% of the mass in molecular clouds, are important because they catalyze the formation of many gas phase molecules, in particular H_2 (Hollenbach & Salpeter 1971) which allow the cooling and collapse of molecular clouds and the formation of stars and planets. High-energy radiation and particles from hot stars, supernovae or active black holes can alter the physical properties of dust grains and thereby affect their role in these processes. Understanding the composition, characteristics, origin and evolution of ISM dust is thus a key question in astrophysics (Draine 2003).

Most of the dust is formed by condensation in the atmospheres of old stars, which eject a significant fraction of their material back into the ISM. Silicates, such as those that produce terrestrial planets, are injected from oxygen-rich asymptotic giant branch (AGB) stars, red supergiants, and supernovae (Tielens 2005). A significant fraction (5-75%) of the silicate dust in stellar winds is observed to be crystalline (Sylvester et al 1999; Molster et al 1999). Specifically, the infrared spectra of the red giants and supergiants reveal the spectroscopic signature of the Mg-rich end members of the olivine and pyroxene families, forsterite (Mg_2SiO_4) and enstatite ($MgSiO_3$). In contrast, the interstellar 9.7 μm silicate feature reveals no evidence for crystalline silicates and, hence, after entering the ISM, crystalline silicates ejected by stellar sources are rapidly amorphized by an unknown mechanism. The upper limit for the crystalline fraction of silicates in the interstellar medium is ~1% (Kemper, Vriend & Tielens 2004; 2005). This conversion of circumstellar crystalline silicates to interstellar amorphous silicates is the *only* solid-state transformation that is observed by astronomers.

Previous laboratory studies have focused on simulating amorphization of complex silicates by low velocity shocks, using irradiation with low energy (keV) ions. Specifically, several experiments using 4-50 keV ions show amorphization (Jaeger et al 2003; Brucato, Strazzulla, Baratta & Colangeli 2004; Demyk et al 2004) for fluences of 10^{15}-10^{17} ions/cm^2. However, this processing is very inefficient, because of the limited range of such low energy ions in solids. Ions of much higher energy, which can penetrate even the largest interstellar grains, pervade the ISM in the form of cosmic rays (>1 MeV/nucleon). However, up till now, processing of dust by such swift ions has been dismissed (Brucato, Strazzulla, Baratta & Colangeli 2004; Watson & Salpeter 1972) because it is believed that the accumulated energy deposition by CR cannot reach the values needed for amorphization (Jaeger et al 2003; Brucato, Strazzulla, Baratta & Colangeli 2004; Demyk et al 2004). In this respect, most of the CR flux consists of H and He ions, but MeV He shows no evidence for amorphization of silicates even for very high ion fluences (Day 1977). However, it is well known that the fluence/energy deposition required for amorphization with keV ions – where elastic atomic interactions dominate – cannot simply be extrapolated to cosmic ray energies - where electronic excitations dominate energy deposition (Ziegler 2003). Indeed, while fast light ions such as H and He do not typically form amorphous tracks in silicates, swift heavy ions (such as Fe) do, a phenomenon that has been studied for decades (Fleischer, Price & Walker 1975) and applied to dating and advanced material modification. Heavy cosmic rays (Fe) are less abundant in the ISM than light cosmic rays (H, He) by a factor of ~10^{-4}. However, at 1 GeV, an Fe ion deposits ~10^4 times more energy per unit path length in silicates (Ziegler 2003) than do protons, and the process of lattice defect formation in this

swift heavy ion irradiation regime is a function of electronic energy loss. Therefore, the cumulative damage of interstellar dust by heavy ion bombardments at high energies can rival or even well exceed that by light ions. Here we demonstrate, using both experiments and modeling, that bombardment by heavy cosmic ray ions is the dominant energetic processing mechanism of dust in the diffuse interstellar medium.

This paper is organized as follows. First, in Section 2, we describe the experimental setup, the choice of the material used, and the methods used to analyze and quantify the results, which are described in Section 3. We discuss our findings in Section 4, which begins with a critical analysis of why CR processing of Galactic ISM dust is indeed important (Section 4.1). For this we estimate the ISM CR spectrum using a 'leaky box' model (Section 4.1.1), and, using phenomenological electronic energy deposition models (Section 4.1.2), we demonstrate that our laboratory experiments at 0.08 MeV/nucleon (but with a large electronic energy loss of ~4.5 keV/nm) can be confidently extrapolated to the relevant (relatively low) CR energies at 1 – 100 MeV/nucleon, where amorphization is most efficient. We then end this section with estimates of the interstellar dust grain amorphization timescale (Section 4.1.3) and the expected fraction of interstellar crystalline silicates. In the subsequent section we compare these results with other possible amorphization processes such as ion bombardment by interstellar shocks (Section 4.2), the lifecycle of crystalline silicates (Section 4.3), and the amorphization of dust grains in the Solar system (Section 4.4) and stellar debris disks (Section 4.5). We then conclude (Section 5) that CR processing of ISM dust is indeed important in our own Galaxy, that it may explain the amorphization of crystalline silicates, and that it is likely to be an even more important effect in forming (starburst) galaxies, which have much

higher cosmic ray fluxes due to 10 – 1000 times larger star formation rates and emerging active black holes (Torres *et al.* 2004).

2. Method

To investigate the effects of cosmic ray impacts on crystalline silicate materials, we have irradiated samples of nominally undoped forsterite (Mg_2SiO_4) single crystals with triply ionized, 10 MeV xenon ions. After irradiation with specific fluences, we have analyzed these samples using several techniques, including Rutherford Backscattering/channeling (RBS/C) spectrometry, infrared (IR) reflection spectroscopy, and cross-sectional transmission electron microscopy (TEM). In this section, we briefly discuss the experimental techniques.

2.1 Ion irradiation and RBS/C spectrometry

The LLNL 4 MV ion accelerator was used for ion bombardment and for RBS/C analysis in single alignment conditions. Samples of laboratory grown undoped forsterite (Mg_2SiO_4) single crystals, (100)-oriented, 10x10x3 mm^3 in size, were bombarded at room temperature with 10 MeV $^{132}Xe^{3+}$ ions over a fluence range of $5x10^{11}$ - $6x10^{13}$ cm^{-2}. Ion incidence was at an angle of ~7° relative to the surface normal to minimize channeling. At this energy, 86% of the total stopping power (energy loss per unit path length) is electronic (4.5 keV/nm, decreasing with penetration depth as the ion slows down [Ziegler 2003]). Furthermore, in this case the xenon ions have a projected range of ~2.4 μms in forsterite (Ziegler, Biersack & Littmark 1985), and, hence can only alter a near-surface layer with thickness close to the projected ion range.

RBS/C (Chu, Mayer & Nicolet 1978) is a well-established powerful technique for the analysis of the elemental composition and crystallinity of the surface and the outer few micrometers of solids. Typically, in RBS/C, a beam of 0.1-3.0 MeV He ions is directed at the sample. The flux and energy distribution of the elastically backscattered particles, measured under a given angle, provides information on the structure of the sample. In our experiments, after irradiation, all samples were characterized by RBS/C with 2 MeV ^{4}He$^+$ ions incident along the [100] direction and backscattered into a detector at 164° relative to the incident beam direction. Damage buildup curves were analyzed with the well-known defect overlap model of cylindrical tracks (Gibbons 1972), where, in the simplest case of so-called zero overlap, the relative concentration of defects is [1-exp(-πR_{track}^2D], where D is ion fluence, and R_{track} is the effective radius of an amorphous track that each impinging ion creates along its path in the crystal. Note that the "bulk" samples used here should require roughly the same amorphization fluence as sub-μm grains (Bringa & Johnson 2004). However, grains smaller than 100 nm may not only amorphize but be destroyed completely by a single CR impact (Bringa & Johnson 2004; Berthelot et al 2000; Toulemonde, Assmann, Gruner & Trautmann 2002).

2.2 Transmission Electron Microscopy – Sample Preparation and Measurements

In TEM, a beam of electrons is focused on the sample. The transmitted electrons form an image, which can be examined for diffraction patterns and, hence, provide information on the structure of the sample. After the ion irradiation experiments, ~100 nm thick forsterite sections were extracted using a FEI 237 Dualbeam focused ion beam (FIB)/field emission scanning electron microscope (FESEM) and FEI Nova 600 dual beam FIB/FESEM

instruments (Graham et al 2004). The extracted cross-sections were attached to the edge of Cu grids using Pt deposition gas in the FIB. The electron transparent FIB sections were examined using a 300 keV Philips CM300 field emission TEM equipped with an Oxford Instruments solid-state energy dispersive X-ray (EDX) spectrometer, EmiSPEC spectral processing software and Digital Micrograph (v. 3.3.1) imaging software. Bright-field and dark-field imaging, electron diffraction and EDX spectroscopy were used to investigate compositions and mineralogy of the extracted sections. All images were recorded using a slow-scan charge-coupled-device (CCD) camera (2048 x 2048 pixels).

2.3 Infrared spectroscopy

Laboratory infrared spectroscopy is a powerful tool to examine the structure of a material. More importantly, it also allows for comparison with astronomical observations. While the infrared spectra of crystalline materials are typically dominated by a few well-defined narrow bands, amorphous materials are characterized by broad and structure-less bands (Dorschner 1999; Jaeger et al 1998). In the present study, we examined the ion-irradiated samples using IR reflection spectroscopy. The infrared measurements were made using an attenuated total infrared reflectance (ATR) single-reflection attachment in a Thermo-Nicolet 670 Nexus Spectrometer operating at 4 cm^{-1} resolution. The beam path and sample compartment were purged continuously with dry air. The ATR used a ZnSe substrate and an angle of incidence of 45$^{\mathrm{o}}$. The reflectance was normalized to the total reflectance of the ZnSe prism without a sample. The sampling depth was less than ~2 μm. Note that, since the index of refraction of crystalline forsterite in the 10-12μm region is even higher than that of ZnSe, the spectra appear different from what would be expected in a transmission experiment. ATR measurements of the samples taken 1 month

after irradiation and repeated ~5-6 months after irradiation produced the same ATR spectra within errors.

3. Results

The astronomical evidence unambiguously demonstrates that crystalline silicates are highly magnesium-rich olivine (forsterite, Mg_2SiO_4) and pyroxene (enstatite, $MgSiO_3$); Sylvester et al. 1999; Molster et al. 2002a,b,c). We have focused on the irradiation effects on forsterite, but enstatite is expected to behave similarly.

The RBS/C spectra of irradiated forsterite samples show a progressive increase in the RBS/C yield with increasing fluence [Fig. 1(a)], which is due to amorphization, revealed by TEM and infrared absorption spectroscopy data discussed below. If we assume that each ion forms a cylindrical track of amorphous material along its path, full amorphization of the near-surface layer is reached as the fluence increases and the tracks overlap. Using the model described above (Gibbons 1972), we derive an effective track radius of (2.8 ± 0.6) nm [Fig. 1(b)].

TEM studies revealed that the sample was amorphized up to a depth nearly half the projected ion range (2400 nm, Ziegler 2003) at a fluence of 3×10^{13}/ions/cm^2, as shown in Fig. 2(a)-(b). TEM results are similar to previous reports for ion-induced amorphization of SiO_2 in the electronic collision regime (Meftah et al. 1994), and of enstatite at the border of electronic and elastic collision regimes (Jaeger et al 2003). Chemical analysis of the amorphous region by energy dispersive X-ray spectroscopy and X-ray photoelectron spectroscopy did not indicate any segregation.

Attenuated total infrared reflectance (ATR) spectra for different ion fluences are shown in Fig. 3(a). The transition from crystalline forsterite – characterized by narrow spectral

features – to an amorphous silicate structure – evidenced by a broad structureless absorption band (Jaeger et al 2003; Brucato, Strazzulla, Baratta & Colangeli 2004; Demyk et al 2004) – occurs at the same fluences as indicated by the RBS/C analysis in Fig. 1.

4. Discussion

4.1 Amorphization by cosmic rays in the interstellar medium

The rate at which crystalline silicates are amorphized in the interstellar medium can be determined by convolving the energy dependent flux of cosmic rays with the "damage" level produced by ions of a given energy. We discuss this in more detail below.

4.1.1 The interstellar cosmic ray spectrum

To estimate the CR flux experienced by interstellar grains, we note that it is not sufficient to consider the high energy CR flux (>1 GeV/nucleon) measured on Earth, since the lower energy CRs – that should dominate amorphization in the ISM – are inhibited from entering the Solar system by the magnetic field in the solar wind. The cosmic ray spectrum can be described using a leaky box model for the propagation and escape of galactic CRs (Ip & Axford 1985). The flux, $\Phi(E)$, as a function of total energy per particle, E, is then given by:

$$\Phi = C\, E^{0.3}/(E + E_o)^3 \quad (cm^2\ s\ sr\ GeV)^{-1} \qquad (Eq.\ 1),$$

where C is a constant that, as we show below, can be determined by a fit to the high energy CR flux measured on Earth, and the scaling factor E_o sets the level of low energy cosmic rays (Webber & Yushak 1983).

Because molecular abundances in interstellar clouds depend ultimately on the cosmic ray

ionization rate, molecular observations provide a direct handle on the low energy cosmic ray flux (Guellin et al. 1977; Wootten et al. 1979; McCall et al. 2003). Observations of OH, HD, and H_3^+ imply a primary cosmic ray ionization rate of about 2×10^{-16} s^{-1} per H-nuclei in the diffuse ISM (Tielens 2005). Using equation 1, the primary cosmic ray ionization rate can be calculated using the differential ionization cross-section (Opal, Beaty & Peterson 1972) fit to the experimental data on H_2 (Rudd, Kim, Madison & Gallagher 1985). The primary CR ionization rate as a function of the scaling factor E_o describing the low energy cosmic ray ionization flux is then:

$$\zeta_p = 5.85 \times 10^{-16} \, (E_o/100 \text{ MeV})^{-2.56} \, s^{-1} \, \text{(H-nuclei)}^{-1}.$$

Hence, the molecular observations imply $E_o \sim 120$ MeV for H.

The value $C = 1.45$ $(cm^2 \text{ s sr GeV}^{-1.7})^{-1}$, was obtained by matching Eq. 1 to experimental data at high energies (e.g. E=1 TeV/particle; Weibel-Sooth, Biermann & Meier 1998) where modulation by the Solar wind has no influence. The high energy data for protons was taken as $\Phi(cm^2 \text{ s sr GeV})^{-1} = \Phi_0 [E(\text{GeV})/(1000 \text{ GeV})]^{-\gamma}$, with $\Phi_0 = 11.51 \, 10^{-9}$ $(cm^2 \text{ s sr GeV})^{-1}$ and $\gamma = 2.77$ (Weibel-Sooth, Biermann & Meier 1998).

The flux of Fe CR responsible for amorphization of interstellar silicates is then provided by Eq. 1, with E_o equal to 56 x 0.12 GeV/particle=6.72 GeV/particle and $C = 8.16 \, 10^{-4}$ $(cm^2 \text{ s sr GeV}^{-1.7})^{-1}$. C was obtained as for the H flux, by matching Eq. 1 to experimental data at E=1 TeV, with $\Phi_0 = 1.78 \, 10^{-9}$ $(cm^2 \text{ s sr GeV})^{-1}$ and $\gamma = 2.6$ (Weibel-Sooth, Biermann & Meier 1998). This gives a Fe flux that is $\sim 5.6 \, 10^{-4}$ smaller than the H flux. Using earlier experimental data, Leger, Jura & Omont (1985) assumed a factor of $\sim 1.6 \, 10^{-4}$ between the H and Fe fluxes, which would give a slightly lower total Fe flux. Fig. 4 shows the resulting H and Fe flux.

4.1.2 Amorphization by ions at interstellar cosmic ray energies

While the ion energies in our experiments (0.08 MeV/nucleon) are in the electronic energy regime, they are lower than relevant cosmic ray energies (1-100 MeV/nucleon). However, based on models for ion damage in solids, validated by experiments on high-energy irradiation of quartz, we can confidently extrapolate our results to the ISM. We have fit the experimental results for several ions bombarding quartz (Meftah et al 1999) to two analytical phenomenological models (Szenes 1997; Tombrello 1994) for track size versus stopping power.

At keV energies, the energy deposition is dominated by elastic atom-atom collisions. At MeV energies and above, corresponding to CR's, energy deposition is mostly due to electronic excitations. Specifically, heavy cosmic rays create nm-sized tracks of dense electronic excitations that can be (partly) converted into atomic motion (Bringa & Johnson 2004; Berthelot et al 2000; Toulemonde, Assmann, Gruner & Trautmann 2002; Szenes 1997; Tombrello 1994; Meftah et al 1994) and can lead to amorphization in a cylinder of the material forming the track. For ion energies typical for CR's, the cross section to create electronic excitations increases with increasing ion mass (Ziegler 2003).

To extrapolate the track size measured for 10 MeV Xe-atoms to astrophysically relevant ions and energies, we have used both the "thermal spike" model proposed by Szenes (1997) and the bond-breaking model by Tombrello (1994). We calculated dE/dx vs. E for both Xe and Fe using the SRIM2003 code (Ziegler 2003) for a forsterite density of 3.2 g/cm^3. Therefore, (dE/dx) and $R_{track}[(dE/dx)]$ are both known as functions of projectile energy. As shown in figure 5, these models, although formulated differently, predict roughly the same size for the amorphous track. The semi-analytic model by Toulemonde

et al (2002) also gives similar results. All these models are expected to give poor results at low dE/dx, close to the threshold for track formation, but still agree within a factor of 2 with the experimental data in this dE/dx range. One can notice the good agreement between these different models and experiments, including our results on forsterite, despite the much higher chemical complexity of forsterite compared to SiO_2. This agreement supports our extrapolation of the track radius to the higher CR energies in the ISM.

4.1.3 The interstellar amorphization timescale

For a given ion energy E, there is a flux $4\pi\Phi(E)$ on each grain. Each ion creates an amorphous track of area πR_{track}^2, which depends on the incident energy through $dE/dx(E)$. The amorphization time, Δt_{amorph}, of a grain can be estimated as:

$$\Delta t_{amorph} = \left(\int_{E_{min}}^{E_{max}} \left\{ \pi R_{track}^2 \left[(dE/dx)(E) \right] \right\} 4\pi\Phi(E) dE \right)^{-1} \qquad \text{(Eq. 2),}$$

It is possible to integrate over multiple CR species, besides Fe, with dE/dx high enough to produce amorphous tracks, but the flux of these other species is negligible and would not significantly change the amorphization time calculated for Fe alone. Eq. 2 assumes that even the less energetic cosmic rays (at E_{min}) will be able to amorphize the ion track all across the grain, which is true for sub-μm grains. In this limit, Δt_{amorph} is independent of the grain area. Eq. 2 neglects any track overlap, but taking into account such track overlap based on a model [Gibbons 1972, Figure 1(b)] further increases the amorphization time by up to a factor of ~4. This is the number we report below. Note that this gives an upper bound for Δt_{amorph} since our calculation assumes that grains are initially 100% crystalline, while only a fraction of them (5%-75%) are in that state

(Sylvester et al 1999; Molster et al 1999).

Note that the minimum kinetic energy for a CR escaping from a supernova remnant shock, E_{min}, can be fixed by requiring that it be at least larger than $2mv^2_{shock}$, where m is the mass of the particle and v_{shock} the shock velocity (Bell 1978). For shock velocities of the order of 10^{3-4} km/s, E_{min} is in the range of a few MeV for a proton, and correspondingly larger for heavier ions. For Fe CR, $E_{min} \sim 0.1$ GeV. Since no tracks form in thin layers for $E > 5$ GeV [$(dE/dx) < 1.5$ keV/nm], we use $E_{max} = 5$ GeV. Using the flux described above gives $\Delta t_{amorph} \sim 70$ million yr. Using $E_{min} = 1$ GeV gives $\Delta t_{amorph} \sim 180$ million yr. These times correspond to ~ 230 and 180 Fe CR impacts every million years on a grain of 0.1 μm radius.

4.1.4 The fraction of interstellar crystalline silicates

Based upon our experiments, we have calculated an exposure time of nearly 70 million yr to completely amorphize crystalline silicates in the ISM. To evaluate quantitatively the crystalline silicate fraction in the ISM, we develop a simple stick model, following McKee (1989) and Kemper, Vriend & Tielens (2004). Silicates are injected into the ISM by stars in the later stages of their evolution in amorphous and crystalline form. Strong shock waves driven by supernova explosions destroy these silicates through sputtering. In addition, dust is lost when new stars are formed. The last process to consider is the amorphization of crystalline grains by cosmic rays. In steady state, the crystalline-to-amorphous mass fraction, δ, is then given by:

$$\delta = (k_{SF} + k_d)\,(k_{CR}\,(1+\delta_o) + k_{SF} + k_d)^{-1}\,\delta_o$$

under the assumption that sputtering destroys crystalline and amorphous silicates at the same rate. Here, k_{SF} is the star formation rate (2×10^{-10} yr^{-1}, corresponding to 1 M_{sun}/yr;

McKee 1989), k_d is the dust destruction rate (2.5×10^{-9} yr^{-1}, corresponding to silicate lifetime of 4×10^8 yr; Jones, Tielens & Hollenbach 1996), k_{CR} is the amorphization rate (1.4×10^{-8} yr^{-1}, corresponding to 7×10^7 yr -this work) and δ_o is the fraction of silicates injected into the ISM in crystalline form. With these values, this equation simplifies to:

$$\delta \sim k_d/k_{CR}\ \delta_o$$

Estimates for δ_o vary from ~0.15 - if AGB stars dominate the dust budget - to ~0.05 if supernovae are important sources of amorphous silicates (Kemper, Vriend & Tielens 2004). The estimated fraction of crystalline silicates ranges then from 0.008 to 0.027, which should be compared to the observed fraction of crystalline silicates of less than 0.01 (Kemper, Vriend & Tielens 2005).

This estimate ignores re-accretion of silicates in the ISM. Studies of interstellar depletion of Si, Mg, and Fe strongly suggest that interstellar grains are covered by a thin layer (~1 nm) of a refractory oxide mantle (Savage & Sembach 1996; Tielens 1998) that is expected to be amorphous due to the low formation temperature. This layer would protect silicates against sputtering in shocks with velocities less than ~150 km/s, which are thought to dominate grain destruction (Savage & Sembach 1996). As a result, the grain destruction rate is likely an order of magnitude smaller and, hence, the estimated crystalline silicate fraction is then less than ~0.003.

4.2 Amorphization by ion bombardment in interstellar shocks

Several studies on amorphization of grains due to keV ions have been reported in the literature, mostly using light (H or He) ions at 4-50 keV energies (Jaeger et al 2003; Bruccato et al 2004; Demyk et al 2004). It was found that crystalline material amorphizes for fluences that are representative of fast shocks in the diffuse ISM (10^{16}-10^{18} (H,

H$^+$)/cm^2; 10^{15}-10^{17} (He, He$^+$)/cm^2). Laboratory infrared spectra of the irradiated, amorphized silicates showed good agreement with observational data. The results suggest that low energy, light ions are efficient in amorphizing and changing the chemical composition of ISM silicate dust grains. However, the volume of a grain affected by keV ion irradiation is to first order set by the range of the ion. To fully amorphize a grain of 0.1 μm radius, H and He ions require an energy of 22 and 32 keV (Ziegler 2003) respectively, corresponding to a ~2,000 km/s shock. In a two-phase ISM, a single supernova remnant processes dust in the equivalent of ~60 M_{SUN} of interstellar gas mass at shock velocities exceeding 1000 km/s (McKee 1989). With a total ISM gas mass of $4.5x10^9$ M_{SUN} and an effective supernova rate of $8x10^{-3}$/yr, the timescale to amorphize is 10^{10} yr (McKee 1989). Therefore, high velocity shocks would not amorphize dust grains efficiently.

4.3 The lifecycle of crystalline silicates

The lifecycle of interstellar dust starts with the nucleation and growth of high temperature condensates such as silicates at high densities and temperatures in the ejecta from stars, such as Asymptotic Giant Branch stars and supergiants. This ejected material is rapidly mixed with other gas and dust in the ISM. A dust grain cycles many times between the intercloud and cloud phases until it is either destroyed by fast (~100 km/s) supernova shocks or is incorporated into newly formed stars or planetary systems. Observations with the Infrared Space Observatory have revealed that crystalline silicates are abundant in the initial stages of this life cycle - the ejecta from stars - as well as in last stages – circumstellar disks around Herbig AeBe stars and T-Tauri stars, but are absent in the intermediate stages (Sylvester et al. 1999; Kemper et al. 2004; 2005; Malfait et al. 1999;

Waters and Waelkens 1998). As our experimental and modeling results demonstrate, crystalline silicates injected into the ISM are rapidly amorphized by galactic cosmic rays on a timescale that is short compared to other relevant timescales (section 4.1.3). The high abundance of crystalline silicates in circumstellar disks surrounding young stars (Waters and Waelkens 1998) must then reflect subsequent processing of grains in these environments. Recent mid-infrared spectroscopic interferometry on AU-scale-sizes has revealed the presence of a strong gradient in the crystallinity of silicates in the circumstellar disks of the three objects investigated (van Boekel et al. 2004). Likely this reflects the rapid annealing in the hot inner regions of these disks coupled with turbulent diffusion outwards of the crystalline grains.

4.4 Amorphization in the Solar system

Crystalline silicates are an abundant component of dust from comets. Likewise, Interplanetary Dust Particles (IDPs) collected in the stratosphere contain silicates (Bradley, Humecki & Germani 1992). It is thus of some interest to consider the amorphization of crystalline silicates in a Solar system setting.

IDPs are part of the zodiacal dust cloud the dust in the inner solar system, a tenuous disk of small (1-200 μm) dust particles that orbits within about 5 AU, fed erratically by the disintegration of comets close to the Sun, and by dust from collisions in the asteroid belt. The lifetimes of small particles in this zodiacal cloud are determined by radiation pressure. We can define β as the ratio of the radiation pressure force to the gravitational attraction by the Sun,

$\beta = F_{rad}/F_{grav} = 6 \times 10^{-1} \, Q_{rp}/a\rho_s$

with Q_r the radiation pressure efficiency, ρ_s the specific density of the material (in g cm^{-3}), and a the grain radius (in μm; de Pater and Lissauer 2001). When β is larger than 0.5, grains on circular orbits are ejected from the Solar system through radiation pressure, while for smaller values of β, grains spiral slowly inwards due to the Poynting-Robertson effect. For grains large compared to the wavelength, the radiation pressure efficiency is approximately unity and $\beta = 0.2/a$ for silicates. The parameter, β, reaches a maximum of about 0.6 for silicate grains of about 0.3 μm and these grains would be blown out. The Poynting-Robertson lifetime, τ_{pr}, for a grain with size a orbiting a star of mass M_* and luminosity L_* at a distance r is given by,

$$\tau_{pr} \sim 400 \ (r/1 \ AU)^2 \ (L_*/L_o) \ (M_o/M_*) \ (1/\beta) = 2000 \ a \ (r/1 \ AU)^2 \ yr$$

Hence, a 100 μm grain is calculated to survive up to 10^6 years in the zodiacal cloud around the Sun. Measurements of galactic and solar cosmic-ray-generated radionuclides have enabled direct estimates of IDP solar system residence times (Bradley et al. 1984; Nishiizumi et al., 1991). These lifetimes are shorter than the timescale against amorphization of silicates by galactic cosmic rays derived in section 4.1.3. In addition, low energy cosmic rays are excluded from the solar system by the Solar wind, further prolonging the lifetime of crystalline silicates against amorphization by cosmic rays. We note that the range of Fe CR in forsterite is ~13 μm at 100 MeV, 100 μm at 750 MeV, and 160 μm at 1 GeV. Assuming a lower energy cut-off of 100 MeV for the interstellar cosmic ray energy, the track density will be constant up to a depth of ~13 μm. Fig. 6 shows the track density as a function of depth.

Interplanetary dust particles will experience a much larger fluence of lower energy, solar wind ions. Adopting the properties of the quiescent Solar wind at 1 AU ($n_p = 5$ cm^{-3}, v =

500 km/s), we calculate a fluence of 7.5×10^{20} protons cm^{-2} over 10^5 yr. A solar wind Fe ion, with ~1 keV/amu has a range of 0.03 μm, while a proton with the same energy/amu has a range of ~0.01 μm. At this energy, even heavy ions will not lead to ionization tracks but to relatively dense collision cascades, and radiation damage will be dominated by solar wind ions only extremely close to the surface, despite their much larger fluence.

4.5 Amorphization of grains in debris disks

The Infrared Astronomical Satellite, IRAS, discovered mid and far-IR emission from main-sequence stars, which is well in excess of the expected photospheric emission (Aumann et al. 1984). This excess emission was attributed to grains resulting from cometesimal and planetesimal collisions in a debris disk surrounding the star. Subsequent studies by the Infrared Space Observatory revealed that more than 15 % of all main sequence have such a debris disk (Habing et al 1999; Lagrange, Backman & Artymowicz 2000). The spectral energy distribution of this excess imply that this emission arises from dust in regions analogous to the Solar system Kuiper belt (i.e., at > 30 AU from the star). Imaging studies have revealed that this emission originates from ring-like structure at such distances (Holland et al. 1998; Greaves et al. 1998; Jayawardhana et al. 1998).

A-type stars do not have stellar winds and, hence, the interstellar CR induced crystallization lifetime calculated in section 4.1.3 can be compared to the dust lifetime in these systems. The lifetime against the Poynting-Robertson effect at R=30 AU from an A star with $M_* = 3$ M_o and $L_* = 80$ L_o is 7×10^4 $a(\mu m)$ yr. These grains will thus not be amorphized by galactic cosmic rays over their lifetime. Grains dynamically trapped in a resonance with an orbiting planet can be much older. The gaps, arcs, rings, warps, clumps, and bright patches observed in images of many debris disks suggests that the

gravitational perturbing effects of embedded planets can be important (Kalas *et al.* 2005). Grains in such long-lived stable structure will be amorphized on a timescale of ~70 million years by galactic cosmic rays and, hence, infrared spectroscopy of this dust may provide therefore a handle on the history and lifetime of such grains. Finally, it should be mentioned that, in some cases, the lifetime of the system may be the limiting factor. For example beta Pic is estimated to have an age of only 12 million years and so amorphization will not have proceeded very far yet even for trapped grains.

5. Conclusion

Our experimental irradiation studies show that crystalline silicates are readily amorphized by bombardment with high-energy ions. This is in agreement with experiments showing amorphization of SiO_2 under heavy ion bombardment, including energies typical of CRs. Using theoretical models we have extrapolated these experimental results to astrophysically relevant energies and demonstrate that cosmic ray ions with energies in the range 0.1-5 GeV will amorphize crystalline silicate grains in the ISM on timescale of 70 million years.

This time is much shorter than the time interval between the injection of silicate materials into the ISM by old stars and their incorporation into new stars and planetary systems (~2500 million yr; Savage & Sembach 1996), and the destruction of silicates in the ISM (400 million yr; Jones, Tielens & Hollenbach 1996). We estimate that, in steady state, less than 0.3% of the crystalline grains injected into the ISM survive amorphization.

The IR spectra of irradiated silicates readily lose the narrow features characteristic of crystalline materials and develop a broad featureless band in the 10-12 µm region [Fig. 3(a)]. This spectral change in reflectance due to amorphization parallels the emission

spectral differences between silicates ejected by old stars and silicate grains observed in the ISM [Fig. 3(b)]. Thus, energetic processing of interstellar silicates by high-energy cosmic ray ions affects the opacity of the ISM.

In addition, we have examined the irradiation of silicates in the solar system and in debris disks. For the Solar system, cosmic ray irradiation of zodiacal dust will have little effect since the Poynting-Robertson effect severely limits the grains lifetime in the inner (5AU) Solar system. Typically, dust is located farther out (~30 AU) in debris disks around A type stars. Nevertheless, amorphization by cosmic rays is still similarly limited by the Poynting-Robertson effect in these environments. We do recognize, however, that cosmic ray processing can play a role for dust trapped in resonances with planetary companions. Infrared spectroscopic studies of such systems may provide a handle on the lifetime of these grains.

We note that other consequences of CR processing are the destruction of small grains (Bringa & Johnson 2004) and the synthesis and ejection of complex molecules formed on the grain surface. CRs will also restructure the grain surfaces, which may affect the formation efficiency of H_2 as suggested by Monte Carlo simulations revealing that the temperature range for high efficiency H_2 formation is larger for heterogeneous than for crystalline surfaces (Cuppen & Herbst 2005). Since we find that the amorphization time scale is so short, nearly all dust grains in the ISM will have been processed and the H_2 and subsequent star formation will be maximized (*positive* feedback).

Energetic processing may have been particularly important in the early universe during the formation and co-evolution of galaxies and their central, galactic black holes (Di Matteo, Springel & Hernquist 2005; Silk & Rees 1998). Those environments are

characterized by both rapid injection of freshly synthesized (crystalline) silicates by short-lived massive stars as well as high supernova rates (e.g., high cosmic ray fluxes) and jets of energetic particles from the central black hole. The crystalline-to-amorphous silicate fraction in these environments is then a balance between the injection and the amorphization process and, based upon our experimental and theoretical studies, observations could then be used to deduce the flux of energetic particles in these environments, which are hard to determine otherwise.

Recently, the spectroscopic signature of crystalline silicate was recognized in the spectra of a sample of deeply embedded Ultra Luminous InfraRed Galaxies (ULIRGs; Spoon et al. 2005). This class of ULIRGs likely reflects a brief, early phase in the evolution of ULIRGs in which star formation and AGN activity has not yet had time to process and disperse the nuclear gas and dust. The presence of copious amounts of crystalline silicates in these starburst environments suggests that the dust has very recently (< 70 Million yrs) been injected – presumably by massive stars formed in the star burst, and therefore has not yet been amorphized by CR ions. This is consistent with ULIRG starburst age estimates of $10 - 100$ Million years (Genzel et al. 1998).

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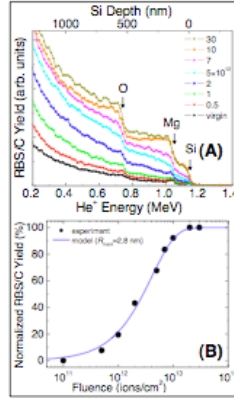
Fig. 1

Fig. 1: (a) Selected RBS/C spectra illustrating the accumulation of lattice disorder in forsterite single crystals irradiated with 10 MeV Xe ions at room temperature at different fluences. The spectrum for a fluence of $3 \times 10^{13}/cm^2$ coincides with the random spectrum. (b) Normalized RBS/C yields fit to a track-overlap amorphization model (Gibbons 1972) giving an effective ion track radius of 2.8 nm.
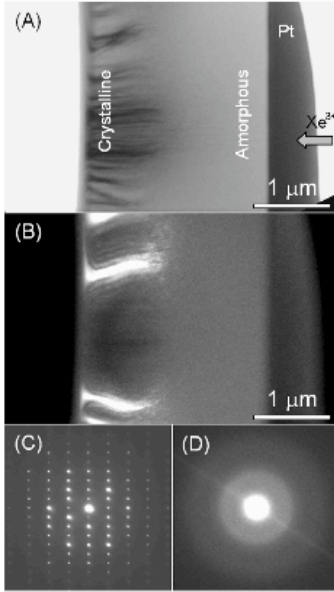
Fig. 2

Fig. 2: Cross-sectional bright-field (a) and dark-field (b) TEM images of single crystal forsterite irradiated at room temperature with 10 MeV Xe ions to a dose of $3 \times 10^{13}/cm^2$. Shown in (c) and (d) are selected-area electron diffraction patterns taken from the left and right parts of the specimen, respectively, confirming the ion-induced structural modification from the initial single crystal to a completely amorphous material.
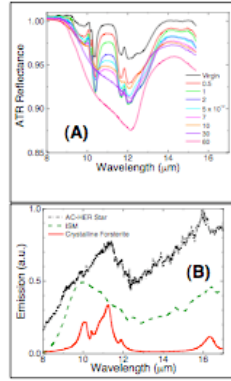
Fig. 3

Fig. 3: (a) ATR infrared spectra of forsterite samples irradiated at different fluences. (b) Astronomical emission spectra showing a broad amorphous band in the ISM (Chiar & Tielens 2005) and crystalline peaks for dust in the ejecta of the red giant, AC Her (Molster, Waters, Tielens & Barlow 2002) A laboratory spectrum of polycrystalline forsterite (Koike et al 2003) is also shown for comparison.
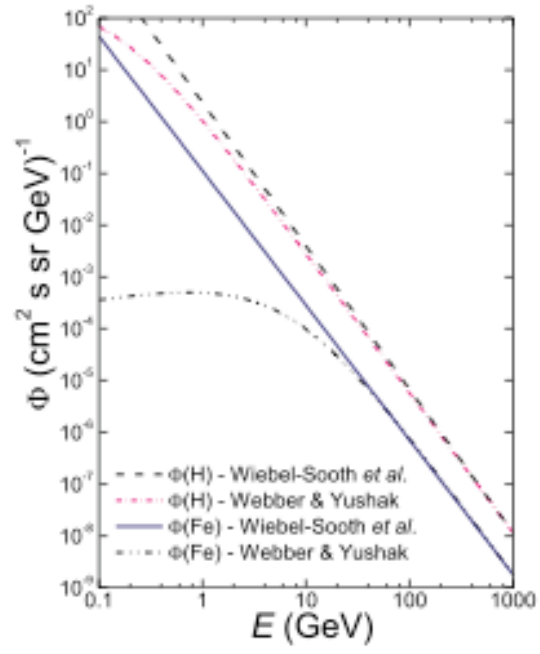
Fig. 4: Cosmic ray flux of H and Fe versus total energy (energy/particle), for both the

unmodulated [Weibel-Sooth et al. 1998] and modulated [Webber&Yushak 1983] case.
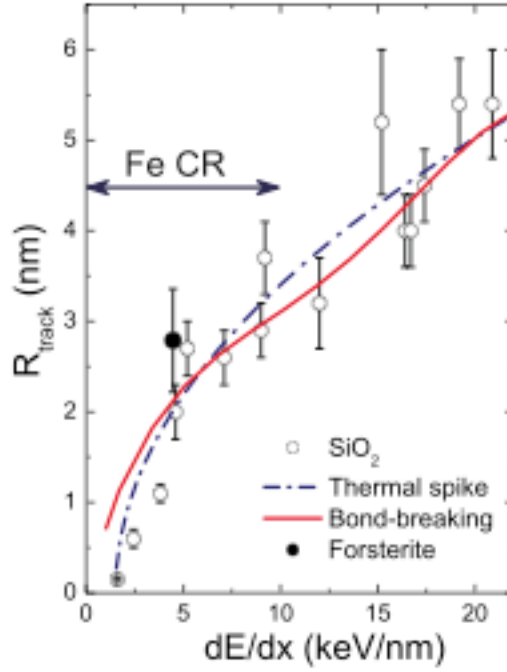
Fig. 5: Amorphous track radius vs. electronic stopping power *dE/dx*. Experimental data points are for forsterite (this work) and quartz (Meftah A. et al. 1994). The lines are results of the thermal spike (Szenes 1997) and bond-breaking (Tombrello 1994) model predictions. The *dE/dx* range for 0.1-10 GeV Fe cosmic rays in forsterite is also shown.
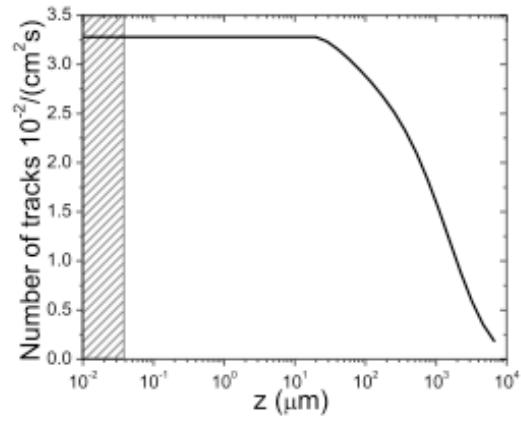
Fig. 6: Track density for Fe CRs with energies larger than 100 MeV as function of depth. The dashed box indicates the range of typical Fe solar wind ions with ~1 keV/amu. Solar flares with higher energy ions would penetrate deeper.